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BULLETIN DE L'ACADÉMIE
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MATHEMATIK

Der Fundamentalsatz über schwache Lösungen der allgemeinen linearen Systeme der elliptischen Differentialgleichungen beliebiger Ordnung

von

K. MAURIN

Eingereicht von K. KURATOWSKI am 28. September 1954

Einführung. In der berühmten Abhandlung von Weyl [1] über die Methode der Orthogonalprojektion spielt eine fundamentale Rolle das sogenannte "Weylsche Lemma", welches bekanntlich besagt, dass die schwache Lösung der Laplace-Gleichung nach einer Korrektur auf der Nullmenge der harmonischen Funktion gleich ist. Dieser Satz wurde in den letzten Jahren auf immer allgemeinere elliptische Differentialgleichungen ausgedehnt und fand seine allgemeinste Formulierung in einer vor kurzem erschienenen Note von F. E. Browder. Browder begnügte sich nicht mit der Verallgemeinerung des Weylschen Lemmas auf allgemeine elliptische Systeme beliebiger Ordnung, sondern vervollständigte es wesentlich, was ihm weitere tiefe Einblicke in die Eigenwertprobleme und die Struktur der Resolvente einer selbstadjungierten Fortsetzung des symmetrischen elliptischen Systems zu machen ermöglichte. Leider ist dieser Satz bis jetzt *ohne* Beweis veröffentlicht. Im Hinblick auf seine grundlegende Rolle habe ich ihn den "Fundamentalsatz über schwache Lösungen der allgemeinen linearen Systeme der elliptischen Differentialgleichungen beliebiger Ordnung" genannt und gebe hier den Beweis einer Verallgemeinerung des Satzes von F. E. Browder. Es gelangt mir nämlich mit Hilfe der wichtigen Untersuchungen von Lopatinskij [3] über Fundamentallösungen der elliptischen Systeme, die Voraussetzungen über die von Browder verlangte Regularität der Koeffizienten abzuschwächen.

Definitionen. Es sei A ein elliptisches System der linearen Differentialoperatoren σ-ter Ordnung mit komplexen Koeffizienten:

$$(Au)_i(x) = \sum_{\nu=1}^{r} \sum_{\substack{\alpha_1+\ldots+\alpha_n=s \\ 0\leqslant s\leqslant\sigma}} a_{\alpha_1\ldots\alpha_s i\nu}(x) \frac{\partial^s}{\partial x_1^{\alpha_1}\ldots\partial x_n^{\alpha_n}} u_\nu(x) \qquad (i,\nu=1,\ldots r).$$

Die Koeffizienten sollen regulär heissen, wenn diejenigen bei den Ableitungen der t-ten Ordnung $\epsilon C^{\sigma+t}(D)$ und die Koeffizienten bei den Ableitungen der σ-ten Ordnung $\epsilon C^{\max(t+2,2\sigma)}(D)$. Die Elliptizität bedeutet folgendes: die Matrix $(a_{ik})_1$ wo

$$a_{i\nu}(x,\xi)=\sum_{\alpha_1,\ldots,\alpha_\sigma=1}^{n} a_{\alpha_1\ldots\alpha_\sigma;\,i\nu}(x)\,\xi_{\alpha_1}\ldots\xi_{\alpha_\sigma} \quad \text{für reelle} \quad \xi_{\alpha_1},\ldots,\xi_{\alpha_\sigma}, \quad \sum_{i=1}^{\sigma}\xi_{\alpha_i}^2\neq 0,$$

ist nichtsingulär für $x\,\epsilon\, D\subset E^n$. Das formal-adjungierte System hat die Form:

$$(A^+(u)_t(x)=\sum_{\nu=1}^{r}\sum_{\substack{\alpha_1+\ldots+\alpha_n=s\\ 0\leqslant s\leqslant\sigma}}(-1)^s\frac{\partial^s\left(\bar a_{\alpha_1\ldots\alpha_s;\nu i}(x)\,u_\nu(x)\right)}{\partial x_1^{\alpha_1}\ldots\partial x_n^{\alpha_n}}.$$

Um das lästige Indices-Schreiben zu vermeiden werden folgende Bezeichnungen benutzt:

$$u(x)=\begin{pmatrix}u_1(x)\\ \vdots\\ u_r(x)\end{pmatrix}; \qquad \omega(z,x)=\left(\omega_{ij}(z,x)\right) \qquad (i,j=1,\ldots,r)$$

$$\eta(z)\,\omega(z,x)=\begin{pmatrix}\sum_{\nu=1}^{r}\eta_\nu(z)\,\omega_{\nu 1}(z,x)\\ \vdots\\ \sum_{\nu=1}^{r}\eta_\nu(z)\,\omega_{\nu r}(z,x)\end{pmatrix}; \qquad \omega(z,x)\,\eta(z)=\begin{pmatrix}\sum_{\nu=1}^{r}\omega_{1\nu}(z,x)\,\eta_\nu(z)\\ \vdots\\ \sum_{\nu=1}^{r}\omega_{r\nu}(z,x)\,\eta_\nu(z)\end{pmatrix}.$$

Das skalare Hilbert-Produkt:

$$(u,v)=\int_D u(x)\,\overline{v(x)}\,dx=\sum_{i=1}\int_D u_i(x)\overline{v_i(x)}\,dx$$

(D-offener Bereich des n-dimensionalen Raumes E^n), wo die Integration in Bezug auf Lebesgue n-Mass ist.

$C^{l,r}(D)$ ist die Menge der r-komponenten Funktionenvektoren, deren Komponenten $\epsilon C^l(D)$.

$K_m(U)$ bedeuten folgende Klassen der $(r\times r)$-Funktionenmatrizen $\varkappa(z,x)=(\varkappa_{ij}(z,x))$ deren Elemente $\varkappa_{ij}(z,x)$ in $U\times U$, (u — ein hinreichend kleines Untergebiet von D), für $x\neq z$ stetig sind,

$$\varkappa_{ij}(z,x)\,|z-x|^m, \quad \text{resp.}\ \frac{\varkappa_{ij}(z,x)}{\lg|z-x|}, \quad \text{resp.}\ \varkappa_{ij}(z,x)$$

gleichmässig beschränkt sind in $U\times U$, für $m>0$, resp. $m=0$, resp. $m<0$.

Bei den über das System gemachten Voraussetzungen gilt der folgende

Hauptsatz: Es sei A ein lineares elliptisches System mit regulären Koeffizienten in $D\subset E^n$. Für jede hinreichend kleine Umgebung $U_\varepsilon\subset D$ des Punktes x gibt es $(r\times r)$-Funktionenmatrizen $r(z,x)$, $\omega(z,x)$ definiert für $(x,z)\,\epsilon\, U_\varepsilon\times U_\varepsilon$, so dass:

a) wenn u für $\eta \in L^{r}(D)$ die schwache Lösung von $Au = \eta$ ist, d. h. $(u, A^{+}\varphi) = (\eta, \varphi)$ für alle $\varphi \in C_C^{l}(D)$ *), dann gelten fast überall in $U_{\varepsilon/2} = \{z \mid |z - x| < \varepsilon/2\}$ folgende Gleichungen:

$$u_i(x) = \sum_{\mu=1}^{r} \int_{U_\varepsilon} u_\nu(z) \overline{r_{\nu i}(z, x)}\, dz + \sum_{\nu=1}^{r} \int_{U_\varepsilon} \eta_\nu(z) \overline{\omega_{i\nu}(z, x)}\, dz \quad (U_\varepsilon = \{z \mid |z - x| < \varepsilon\}).$$

b) bei den Voraussetzungen aus a), $u \in C^{l,r}(D)$ nach der Korrektur auf einer Nullmenge für $l < \sigma - n/2$.

c) wenn dazu noch η einer Hölderbedingung genügt, dann ist nach der Korrektur auf einer Nullmenge, $u \in C^{\sigma,r}(D)$ und $Au(x) = \eta(x)$.

d) $r_{ij}(z, x)$ sind σ-mal stetig nach x in $U_\varepsilon \times U_\varepsilon$ differenzierbar; und

$$\frac{\partial^{\alpha_1+\ldots+\alpha_n+\beta_1+\ldots+\beta_n} \omega_{ij}(z, x)}{\partial x^{\alpha_1} \ldots \partial x^{\alpha_n} \partial z^{\beta_1} \ldots \partial z_n^{\beta_n}} \in K_{n+\alpha_1+\ldots+\alpha_n+\beta_1+\ldots+\beta_n-\sigma}(U_\varepsilon).$$

Bevor wir an den Beweis des Hauptsatzes herantreten, soll das wichtige Ergebnis von Lopatinskij in der hier nötigen Form wiedergegeben werden:

Lemma ([3] S. 311, 313). In jeder hinreichend kleinen Umgebung $U \subset D$ des Punktes x, gibt es eine $(r \times r)$-Funktionenmatrix $\omega(z, x)$ (Fundamentallösung des Systems $A^{+}u = 0$) mit folgenden Eigenschaften:

1^0 es gilt:

$$\frac{\partial^{\alpha_1+\ldots+\alpha_n+\beta_1+\ldots+\beta_n}}{\partial x_1^{\alpha_1} \ldots \partial x_n^{\alpha_n} \partial z_1^{\beta_1} \ldots \partial z_n^{\beta_n}} \omega(z, x) \in K_{n+\alpha_1+\ldots+\alpha_n+\beta_1+\ldots+\beta_n-\sigma}(U); \tag{1}$$

2^0 für jede $\chi(z) \in C^{l,r}(D)$ gilt:

$$A_z^{+} \int_V \omega(z, x)\, \chi(x)\, dx = \mathcal{E}(z)\, \chi(z), \tag{2}$$

wo V solcher beliebiger offener Bereich ist, dass $\overline{V} \subset U$, und

$$\mathcal{E}(z) = \begin{cases} 1 & \text{für } x \in V \\ 0 & \text{für } x \notin V. \end{cases}$$

Beweis des Hauptsatzes:

ad a) und d)

Da der Hauptsatz offenbar einen lokalen Charakter hat, genügt es ihm für die hinreichend kleine Kugel U_ε zu beweisen. Sei nämlich $U_\varepsilon = \{y \mid |y - x| < \varepsilon\}$, wobei $\varepsilon > 0$ so gewählt wurde, damit $U_\varepsilon \subset D$ und für U_ε das Lemma gelte. Es sei $\varrho_\varepsilon(r)$ eine solche Funktion aus $C^{\infty}(0, \infty)$ so dass

$$\varrho_\varepsilon(r) = \begin{cases} 1 & \text{für } 0 \leqslant r \leqslant \varepsilon/4 \\ 0 & \text{für } r \geqslant \varepsilon/2. \end{cases}$$

*) $\varphi \in C_c^{l}(V)$ soll bedeuten, dass $\varphi \in C^{l}(V)$ und einen kompakten Träger $\subset V$ hat.

Da $u(x)$ die schwache Lösung von $Au = \eta$ ist, haben wir:

$$\int_{U_\varepsilon} \eta(z)\,\overline{\Psi(z)}\,dz = \int_{U_\varepsilon} u(z)\,\overline{A_z^+ \Psi(z)}\,dz \quad \text{für} \quad \Psi \in C_C^{\sigma,r}(U_\varepsilon). \tag{3}$$

Wir setzen

$$\begin{aligned} \Psi(z) &= \int_{U_{\varepsilon/2}} \varrho_\varepsilon(|t-z|)\,\omega(z,t)\,\varphi(t)\,dt \\ &= \int_{U_{\varepsilon/2}} [\varrho_\varepsilon(|t-z|) - 1]\,\omega(z,t)\,\varphi(t)\,dt + \int_{U_{\varepsilon/2}} \omega(z,t)\,\varphi(t)\,dt, \end{aligned} \tag{4}$$

wobei $\varphi \in C_C^{l,r}(U_{\varepsilon/2})$. Daher ist nach (1), (2):

$$A_z^+ \Psi(z) = \mathcal{E}(z)\,\varphi(z) + \int_{U_\varepsilon} A_z^+ \{[\varrho_\varepsilon(|t-z|) - 1]\,\omega(z,t)\}\,\varphi(t)\,dt. \tag{5}$$

Aus (3), (4) haben wir:

$$\int_{U_\varepsilon} \eta(z)\,\overline{\Psi(z)}\,dz = \int_{U_{\varepsilon/2}} \varphi(t) \left\{ \int_{U_\varepsilon} \eta(z)\,\overline{\varrho_\varepsilon(|x-z|)\,\omega(z,t)}\,dz \right\} dt$$

und wegen (3), (5):

$$\int_{U_\varepsilon} \eta(z)\,\overline{\Psi}(z)\,dz = \int_{U_\varepsilon} u(z) \left\{ \overline{\mathcal{E}(z)\,\varphi(z)} - \overline{\int_{U_{\varepsilon/2}} r(z,t)\,\varphi(t)\,dt} \right\} dz,$$

wobei $A_z^+ \{[\varrho_\varepsilon(|t-z|) - 1]\,\omega(z,t)\} = -r(z,t)$ gesetzt wurde.

Schliesslich:

$$\int_{U_{\varepsilon/2}} u(t)\,\overline{\varphi(t)}\,dt = \int_{U_{\varepsilon/2}} \overline{\varphi(t)} \left\{ \int_{U_\varepsilon} \eta(z)\,\varrho_\varepsilon(|t-z|)\,\omega(z,t)\,dz + \int_{U_\varepsilon} u(z)\,r(z,t)\,dz \right\} dt.$$

Also

$$u(x) = \int_{U_\varepsilon} u(z)\,\overline{r(z,x)}\,dz + \int_{U_\varepsilon} \eta(z)\,\overline{\underset{1}{\omega}(z,x)}\,dz,$$

fast überall in $U_{\varepsilon/2}$, wobei $\underset{1}{\omega}(z,x) = \varrho_\varepsilon(|x-z|)\,\omega(z,x)$ gesetzt wurde. Wegen (1) ist zugleich d) erwiesen worden w. z. b. w.

ad b)

Es genügt offenbar zu zeigen, dass

$$g(z) = \int_{U_\varepsilon} \eta(z)\,\overline{\underset{1}{\omega}(z,x)}\,dz \in C^{l,r}(u_\varepsilon) \qquad l < \sigma - n/2.$$

Wir setzen:

$$g^{\alpha_1 \ldots \alpha_n}(x) = \int_{U_\varepsilon} \eta(z)\,\frac{\partial^{\alpha_1+\ldots+\alpha_n}}{\partial x_1^{\alpha_1} \ldots \partial x_n^{\alpha_n}}\,\overline{\underset{1}{\omega}(z,x)}\,dz;$$

$$g_\delta^{\alpha_1 \ldots \alpha_n}(x) = \int_{U_\varepsilon} \eta(z)[1 - \varrho_\delta(|x-z|)]\,\frac{\partial^{\alpha_1+\ldots+\alpha_n}}{\partial x_1^{\alpha_1} \ldots \partial x_n^{\alpha_n}}\,\overline{\underset{1}{\omega}(z,x)}\,dz;$$

aber

$$\left|\int_{U_\varepsilon} \eta(z)\varrho_\delta(|x-z|)\frac{\partial^{\alpha_1+\ldots+\alpha_n}}{\partial x_1^{\alpha_1}\ldots\partial x_n^{\alpha_n}}\omega_1(|x-z|)\,dz\right| \leqslant \|\eta\| L_\delta^{\alpha_1\ldots\alpha_n},$$

für $n+\alpha_1+\ldots+\alpha_n-\sigma<n/2$

gleichmässig in $x \in U_\varepsilon$ wegen dem Lemma und der Schwarzschen Ungleichung; also $\left|g^{\alpha_1\ldots\alpha_n}(x)-g_\delta^{\alpha_1\ldots\alpha_n}(x)\right| \leqslant \|\eta\| L_\delta^{\alpha_1\ldots\alpha_n}$.

Da $\lim_{\varepsilon\to 0} L_\delta^{\alpha_1\ldots\alpha_n}=0$, haben wir $g^{\alpha_1\ldots\alpha_n}(x)=\lim_{\delta\to 0} g_\delta^{\alpha_1\ldots\alpha_n}(x)$ gleichmässig in $x \in U_\varepsilon$. Da

$g_\delta^{\alpha_1\ldots\alpha_n}(x) \in C^{0,r}(U_\varepsilon)$, ist $g^{\alpha_1\ldots\alpha_n}(x) \in C^{0,r}(U_\varepsilon)$, wenn $\alpha_1+\ldots+\alpha_n<\sigma-n/2$:

dass

$$\frac{\partial^{\alpha_1+\ldots+\alpha_n}}{\partial x_1^{\alpha_1}\ldots\partial x_n^{\alpha_n}}g(x)=\int_{U_\varepsilon}\eta(x)\frac{\partial^{\alpha_1+\ldots+\alpha_n}}{\partial x_1^{\alpha_1}\ldots\partial x_n^{\alpha_n}}\overline{\omega_1(z,x)}\,dz$$

beweist man jetzt wörtlich wie Bochner ([4] S. 343—4)

ad c)

$\eta(z)$ soll jetzt einer Hölderbedingung mit dem Koeffizienten $0<\alpha\leqslant 1$ genügen. Wegen Lemma und der klassischen Sätze über die Integrale vom Potentialtypus ([5] S. 203—4) haben wir $g(x) \in C^{\sigma,r}(U_\varepsilon)$.

Hiermit ist der Beweis des Hauptzatzes vollständig erbracht.

Mathematisches Institut der Polnischen Akademie der Wissenschaften

SCHRIFTTUM

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MATHÉMATIQUE

Sur quelques propriétés des fonctions $\varphi(n)$ et $\sigma(n)$

par

A. SCHINZEL et W. SIERPIŃSKI

Présenté par W. SIERPIŃSKI le 4 Octobre 1954

Le but de cette communication et de démontrer d'une façon élémentaire (sans faire appel au théorème de Lejeune-Dirichlet sur la progression arithmétique) quatre théorèmes concernant les fonctions $\varphi(n)$ (nombre de nombres naturels premiers avec n et ne dépassant pas n) et $\sigma(n)$ (somme de tous les diviseurs naturels de n).

Théorème 1. *Pour tout nombre naturel m, il existe un nombre naturel $n>1$ tel que $\varphi(n-1)/\varphi(n)>m$ et $\varphi(n+1)/\varphi(n)>m$.*

Démonstration. Soit p_i le i-ème nombre premier. On démontre d'une façon élémentaire [1], que, étant donné un nombre naturel m, il existe un nombre naturel k, tel que

$$(1) \qquad \left(1-\frac{1}{p_1}\right)\left(1-\frac{1}{p_2}\right)\dots\left(1-\frac{1}{p_k}\right)<\frac{1}{4m}.$$

Posons $n=p_1 p_2 \dots p_k$. Tout diviseur premier d du nombre $n-1$, de même que celui du nombre $n+1$, diffère, comme on le voit sans peine, de p_i pour $i=1,2,\dots,k$; donc $d>p_k$. Par conséquent, si $n\pm 1 = q^{\alpha_1}, q^{\alpha_2} \dots q^{\alpha_s}$ est le développement du nombre $n\pm 1$ en facteurs premiers, où $q_1<q_2<\dots<q_s$, on a $q_1>p_k$. En supposant que $s>k$, donc que $s\geqslant k+1$, on aurait $n\pm 1\geqslant q_1 q_2 \dots q_s > p_k^{k+1}>n$, ce qui est impossible, car il n'y a aucun entier intermédiaire entre n et $n+1$. On a donc $s\leqslant k$.

On a en outre d'après une formule connue sur la fonction $\varphi(n)$

$$(2) \qquad \frac{\varphi(n)}{n}=\left(1-\frac{1}{p_1}\right)\left(1-\frac{1}{p_2}\right)\dots\left(1-\frac{1}{p_k}\right) \quad \text{et} \quad \frac{\varphi(n\pm 1)}{n\pm 1}=\left(1-\frac{1}{q_1}\right)\left(1-\frac{1}{q_2}\right)\dots\left(1-\frac{1}{q_s}\right)$$

Comme $q_j>p_k$ pour $j=1,2,\dots,s$, on voit aisément que $s\leqslant k$ et $p_k>k$

$$\prod_{j=1}^{s}\left(1-\frac{1}{q_j}\right)\geqslant\prod_{i=1}^{s}\left(1-\frac{1}{p_k+j}\right)=\frac{p_k}{p_k+s}\geqslant\frac{p_k}{p_k+k}>\frac{1}{2}.$$

D'après (2) et (1) on a donc

$$\frac{\varphi(n\pm1)}{n\pm1}:\frac{\varphi(n)}{n}>2m, \quad \text{d'où} \quad \varphi(n\pm1)/\varphi(n)>2m\frac{n\pm1}{n}>m.$$

Le théorème 1 se trouve ainsi démontré.

Théorème 2. *Pour tout nombre naturel m, il existe un nombre naturel $n>1$ tel que $\varphi(n)/\varphi(n-1)>m$ et $\varphi(n)/\varphi(n+1)>m$.*

Démonstration. Étant donné un nombre naturel m, il existe des nombres naturels k et l tels que:

$$\left(1-\frac{1}{p_2}\right)\left(1-\frac{1}{p_2}\right)\dots\left(1-\frac{1}{p_k}\right)<\frac{1}{4m},$$

$$\left(1-\frac{1}{p_{k+1}}\right)\left(1-\frac{1}{p_{k+2}}\right)\dots\left(1-\frac{1}{p_{k+l}}\right)<\frac{1}{2m}. \tag{3}$$

Posons $a=p_1p_2\dots p_k$ et $b=p_{k+1}p_{k+2}\dots p_{k+l}$. On a donc $(a,b)=1$ et il existe, comme on sait, des entiers x et y tels que $ax-by=1$. Posons $u=x-bE\frac{x}{b}$ et $v=y-aE\frac{x}{b}$ (le symbole Et désignant le plus grand entier ne dépassant pas t). On a $au-bv=1$ et, comme $\frac{x}{b}-1<E\frac{x}{b}\leqslant\frac{x}{b}$, on constate que $0\leqslant u<b$. En supposant que $u=0$, on aurait $-bv=1$ ce qui est impossible, puisque $b>1$. On a donc $u<b$.

Posons $n=2au-1$. C'est donc un nombre naturel et on a $n>1$. Comme $au=bv+1$, on a aussi $n=2bv+1$. Enfin, comme $u<b$, on a $n=2au-1<2ab-1$.

Soit $n=q_1^{\alpha_1}q_2^{\alpha_2}\dots q_s^{\alpha_s}$, où $q_1<q_2<\dots<q_s$, le développement de n en facteurs premiers. Vu l'égalité $n=2au-1=2bv+1$ et les développements de a et de b, on conclut que $q_1>p_{k+1}\geqslant k+1+1$. Or, $n<2ab=$ $=2p_1p_2\dots p_{k+l}<p_{k+l}^{k+l+1}$ et $n\geqslant q_1^s>p_{k+l}^s$ d'où $s<k+l+1$, donc $s\leqslant k+l$.

Comme $q_j>p_{k+1}$ pour $j=1,2,\dots,s$ on a $q_j\geqslant p_{k+1}+j$ pour $j=1,2,\dots s$, donc, vu que $s\leqslant k+l$ et $p_{k+1}>k+l$,

$$\prod_{j=1}^{s}\left(1-\frac{1}{q_j}\right)\geqslant\prod_{j=1}^{s}\left(1-\frac{1}{p_{k+l}+j}\right)=\frac{p_{k+l}}{p_{k+l}+s}\geqslant\frac{p_{k+l}}{p_{k+l}+k+l}>\frac{1}{2}.$$

Par conséquent,

$$\frac{\varphi(n)}{n}=\prod_{j=1}^{s}\left(1-\frac{1}{q_j}\right)>\frac{1}{2}. \tag{4}$$

L'égalité $n=2au-1=2bv+1$ entraîne en outre $a|n+1$ et $b|n-1$, d'où $p_1p_2\dots p_k|n+1$ et $p_{k+1}p_{k+2}\dots p_{k+l}|n-1$. On a donc d'après (3):

$$\frac{\varphi(n+1)}{n+1}\leqslant\left(1-\frac{1}{p_1}\right)\left(1-\frac{1}{p_2}\right)\dots\left(1-\frac{1}{p_k}\right)<\frac{1}{4m}$$

et

$$\frac{\varphi(n-1)}{n-1}\leqslant\left(1-\frac{1}{p_{k+1}}\right)\dots\left(1-\frac{1}{p_{k+l}}\right)<\frac{1}{2m},$$

d'où, en vertu de (4)

$$\frac{\varphi(n)}{\varphi(n+1)}=\frac{\varphi(n)}{n}\cdot\frac{n+1}{\varphi(n+1)}\cdot\frac{n}{n+1}>2m\frac{n}{n+1}>m$$

$\left(\text{puisque } n>1 \text{ et } 2\frac{n}{n+1}>1\right)$ et

$$\frac{\varphi(n)}{\varphi(n-1)}=\frac{\varphi(n)}{n}\cdot\frac{n-1}{\varphi(n-1)}\cdot\frac{n}{n-1}>m.$$

Le théorème 2 se trouve ainsi démontré.

Comme corollaire immédiat du théorème 1 (ou bien du théorème 2), on obtient les formules suivantes, établies par B. S. K. R. Somayajulu [2]:

$$\overline{\lim_{n\to\infty}}\frac{\varphi(n+1}{\varphi(n)}=+\infty \quad \text{et} \quad \underline{\lim_{n\to\infty}}\frac{\varphi(n+1)}{\varphi(n)}=0.$$

Lemme. *Pour tout nombre naturel $n>1$, on a*

$$\frac{n^2}{2}<\varphi(n)\,\sigma(n)<n^2.$$

Démonstration. Si le développement de n en facteurs premiers est $n=q_1^{\alpha_1}q_2^{\alpha_2}\dots q_k^{\alpha_k}$, on a

$$\varphi(n)\,\sigma(n)=n^2\left(1-\frac{1}{q_2^{\alpha_1+1}}\right)\left(1-\frac{1}{q_2^{\alpha_2+1}}\right)\dots\left(1-\frac{1}{q_k^{\alpha_k+1}}\right),$$

d'où $\varphi(n)\,\sigma(n)<n^2$.

D'autre part,

$$\left(1-\frac{1}{q_1^{\alpha_1+1}}\right)\left(1-\frac{1}{q_2^{\alpha_2+1}}\right)\dots\left(1-\frac{1}{q_k^{\alpha_k+1}}\right)\geqslant$$

$$\geqslant\left(1-\frac{1}{2^2}\right)\left(1-\frac{1}{3^2}\right)\left(1-\frac{1}{4^2}\right)\dots\left(1-\frac{1}{(k+1)^2}\right)=\frac{k+2}{2(k+1)}>\frac{1}{2},$$

d'où $\varphi(n)\,\sigma(n)>\frac{n^2}{2}$. Le lemme est ainsi démontré.

Théorème 3. *Pour tout nombre naturel m, il existe un nombre naturel $n>1$ tel que $\sigma(n-1)/\sigma(n)>m$ et $\sigma(n+1)/\sigma(n)>m$.*

Démonstration. D'après le théorème 2, il existe pour tout m naturel un $n>1$ naturel, tel que $\varphi(n)/\varphi(n-1)>8m$ et $\varphi(n)/\varphi(n+1)>8m$. Comme

$$\varphi(n)\,\sigma(n)>n^2,\ \varphi(n-1)\,\sigma(n-1)>\frac{(n-1)^2}{2} \quad \text{et} \quad \varphi(n+1)\,\sigma(n+1)>\frac{(n+1)^2}{2},$$

en vertu du lemme on conclut que

$$\frac{\sigma(n-1)}{\sigma(n)}>\frac{(n-1)^2}{2n^2}\,\frac{\varphi(n)}{\varphi(n-1)}\,\frac{1}{8}\cdot\frac{\varphi(n)}{\varphi(n-1)}>m$$

et

$$\frac{\sigma(n+1)}{\sigma(n)}>\frac{(n+1)^2}{2n^2}\,\frac{\varphi(n)}{\varphi(n+1)}>4m,$$

puisque $\frac{n-1}{n}>\frac{1}{2}$ pour $n>1$. Le théorème 3 est ainsi démontré.

Théorème 4. *Pour tout nombre naturel m, il existe un nombre naturel $n>1$, tel que $\sigma(n)/\sigma(n-1)>m$ et $\sigma(n)/\sigma(n+1)>m$.*

Démonstration. D'après le théorème 1, il existe pour tout m un $n>1$ naturel, tel que $\varphi(n-1)\varphi/(n)>8m$ et $\varphi(n+1)/\varphi(n)>8m$. Comme

$$\varphi(n)\,\sigma(n)>\frac{n^2}{2},\qquad \varphi(n-1)\,\sigma(n-1)\leqslant(n-1)^2,\qquad \varphi(n+1)\,\sigma(n+1)<(n+1)^2$$

en vertu du lemme, on conclut que

$$\frac{\sigma(n)}{\sigma(n-1)}>\frac{n^2}{2(n-1)^2}\,\frac{\varphi(n-1)}{\varphi(n)}>4m \quad\text{et}\quad \frac{\sigma(n)}{\sigma(n+1)}>\frac{n^2}{2(n+1)^2}\,\frac{\varphi(n+1)}{\varphi(n)}>m,$$

puisque $\dfrac{n}{n+1}>\dfrac{1}{2}$ pour $n>1$. Le théorème 4 est ainsi démontré.

Dans le même ordre d'idées, A. Schinzel a établi récemment les propriétés suivantes de la fonction $\varphi(n)$ et les propriétés analogues de la fonction $\sigma(n)$ [3]:

Quels que soient les nombres naturels m et $k>1$, il existe un nombre naturel n, tel que

$$\frac{\varphi(n+i)}{\varphi(n+i-1)}>m \qquad \text{pour} \quad i=1,2,\ldots,k,$$

de même qu'un nombre naturel n, tel que

$$\frac{\varphi(n+i-1)}{\varphi(n+i)}>n \qquad \text{pour} \quad i=1,2,\ldots,k.$$

Les démonstrations de ces théorèmes font l'objet de la communication suivante.

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MATHÉMATIQUE

Quelques théorèmes sur les fonctions $\varphi(n)$ et $\sigma(n)$

par

A. SCHINZEL

Présenté par W. SIERPIŃSKI le 4 Octobre 1954

La communication présente a pour but — de même que la communication précédente — de démontrer d'une façon élémentaire quelques théorèmes sur les fonctions numériques $\varphi(n)$ et $\sigma(n)$. Ces théorèmes constituent des solutions affirmatives des problèmes qui ont été proposés par W. Sierpiński.

Théorème 1. *Deux nombres naturels m et $k>1$ étant donnés, il existe un nombre naturel n tel que*

$$\frac{\varphi(n+i)}{\varphi(n+i-1)}>m \quad \textit{pour} \quad i=1,2,\dots k. \tag{1}$$

Démonstration. D'après un théorème bien connu de la théorie des congruences, $a_1, a_2, \dots, a_k$ étant une suite finie quelconque de nombres naturels deux à deux premiers entre eux, il existe un entier x tel que $x \equiv i \pmod{a_i}$ pour $i=1,2,\dots,k$ et, x_0 étant un tel entier, $x=x_0+ta_1a_2\dots a_k$, où $t=0,\pm 1,\pm 2,\dots$, l'est aussi. On peut donc déterminer l'entier t de manière que $0\leqslant x<a_1a_2\dots a_k$. Or, si $a_1\geqslant k>1$ et $a_2>1$, il résulte de $x\equiv 1 \pmod{a_1}$ et de $x\equiv 2 \pmod{a_2}$ que $x>a_1$ et $x>k$.

Soit c le plus petit des nombres $\varphi(i)/i$, où $i=1,2,\dots,k-1$. Comme $\varphi(1)/1=1$, on a $c\leqslant 1$.

Soit p_i le i-ème nombre premier. Le produit infini $\prod_{i=1}^{\infty}\left(1-\frac{1}{p_i}\right)$ divergeant, comme on sait, vers 0, on peut définir par l'induction les nombres $a_1, a_2, \dots, a_k$ et $l_0<l_1<\dots<l_k$ de façon à avoir

$$a_0=1, \qquad a_i=p_{l_{i-1}+1}\,p_{l_{i-1}+2}\dots p_{l_i} \quad \text{pour } i=1,2,\dots,k,$$

$$l_0=0, \qquad p_{l_1}>k, \qquad \frac{\varphi a_{(i+1)}}{a_{i+1}}<\frac{c}{2m}\,\frac{\varphi(a_i)}{a_i} \quad \text{pour } i=0,1,\dots,k-1.$$

Comme $a_1 = p_1 p_2 \dots p_{l_1} \geqslant p_{l_1} \geqslant k$ et $a_2 > 1$, il existe un entier x tel que $k < x < a_1 a_2 \dots a_k$ et $x \equiv i \pmod{a_i}$ pour $i = 1, 2, \dots, k$.

Le nombre $n = x - k$ est donc un nombre naturel et on a

$$n + k < a_1 a_2 \dots a_k \quad \text{et} \quad a_i \mid n + k - 1 \quad \text{pour} \quad i = 1, 2, \dots, k. \tag{2}$$

On a donc pour $i, j = 1, 2, \dots, k$

$$(n + k - i, a_j) = (n + k - i - (n + k - j), a_j) = (i - j, a_j) = 1 \text{ pour } 1 < j \neq i, \tag{3}$$

puisque $|i - j| < k \leqslant p_{l_1} < a_j$ pour $1 < j \neq i$, et

$$(n + k - i, a_1) = (n + k - i - (n + k - 1), a_1) = (i - 1, a_1) < d_i \leqslant |i - 1| \tag{4}$$

pour $i \neq 1$, enfin

$$(n + k,\ p_1, p_2 \dots p_{l_k}) = (n + k, a_1) = (n + k - (n + k - 1), a_1) = (1, a_1) = 1.$$

Le nombre $n + k$ a donc le développement en facteurs premiers

$$n + k = q_1^{\beta_1} q_1^{\beta_2} \dots q_s^{\beta_s}, \quad \text{où} \quad p_{l_k} < q_1 < q_2 < \dots q_s \quad \text{et} \quad s < k$$

d'après (2).

On a aussi les développements

$$n + k - 1 = p_1^{\alpha_1} p_2^{\alpha_2} \dots p_{l_1}^{\alpha_1} q_1'^{\beta_1'} q_2'^{\beta_2'} \dots q_s'^{\beta_s'}, \quad \text{où} \quad p_{l_k} < q_1' < \dots < q_s'$$

et, d'après (3) et (4), pour $i > 1$

$$n + k - i = d_i p_{l_{i-1}+1}^{\alpha_{l_{i-1}+1}^{(i)}} p_{l_{i-1}+2}^{\alpha_{l_{i-1}+2}^{(i)}} \dots p_{l_i}^{\alpha_{l_i}^{i}} q_1^{(i)\beta_1^{(i)}} \dots q_{s^{(i)}}^{(i)\beta_{s^{(i)}}^{(i)}}$$

où $p_{l_k} < q_1^{(i)} < \dots < q_{s^{(i)}}^{(i)}$.

Évidemment,

$$\frac{\varphi(q_1^{\beta_1} q_2^{\beta_2} \dots q_s'^{\beta_s})}{q_1^{\beta_1} q_2^{\beta_2} \dots q_s^{\beta_s}} = \left(1 - \frac{1}{q_1}\right)\left(1 - \frac{1}{q_2}\right) \dots \left(1 - \frac{1}{q_s}\right)$$

$$\geqslant \left(1 - \frac{1}{p_{l_k} + 1}\right)\left(1 - \frac{1}{p_{l_k} + 2}\right) \dots \left(1 - \frac{1}{p_{l_k} + s}\right) = \frac{p_{l_k}}{p_{l_k} + s} > \frac{1}{2},$$

puisque $s < k < p_{l_k}$.

On trouve pareillement l'inégalité

$$\frac{\varphi(q_1^{(i)\beta_1^{(i)}} q_2^{(i)\beta_2^{(i)}} \dots q_{s^{(i)}}^{(i)\beta_{s^{(i)}}^{(i)}})}{q_1^{(i)\beta_1^{(i)}} q_2^{(i)\beta_2^{(i)}} \dots q_{s^{(i)}}^{(i)\beta_{s^{(i)}}^{(i)}}} > \frac{1}{2}.$$

On a donc

$$\frac{\varphi(n + k)}{n + k} > \frac{1}{2}, \qquad \frac{\varphi(n + k - 1)}{n + k - 1} > \frac{\varphi(a_1)}{2},$$

$$\frac{\varphi(n + k - i)}{n + k - i} > \frac{\varphi(d_i)}{d_i} \frac{\varphi(a_i)}{a_i} \frac{1}{2} \geqslant \frac{c}{2} \frac{\varphi(a_i)}{a_i} \quad \text{pour} \quad 1 < i < k.$$

D'autre part, comme $a_i \mid n+k-i$ pour $i=1,2,\ldots,k$, d'après (2) on a évidemment

$$\frac{\varphi(n+k-i-1)}{n+k-i-1} < \frac{\varphi(a_i+1)}{a_{i+1}} \quad \text{pour} \quad k > i \geqslant 0.$$

Par conséquent,

$$\frac{\varphi(n+k)}{\varphi(n+k-1)} > \frac{1}{2}\frac{\varphi(a_1)}{a_1} > \frac{m}{c} \geqslant m$$

et

$$\frac{\varphi(n+k-i)}{\varphi(n+k-i-1)} > \frac{c}{2}\frac{\varphi(a_1)}{\varphi(a_{i+1})}\frac{a_{i+1}}{a_i}\frac{n+n-i}{n+k-i-1} > m,$$

pour $i=0,1,\ldots,k-1$,

Le théorème 1 se trouve ainsi démontré.

Théorème 2. *Deux nombres naturels m et $k>1$ étant donnés, il existe un nombre naturel n tel que*

$$\frac{\varphi(n+i)}{\varphi(n+i+1)} > m \quad \textit{pour} \quad i=0,1,2,\ldots,k-1.$$

La démonstration est tout à fait analogue à celle du théorème 1, sauf que les nombres a_i doivent satisfaire aux conditions

$$\frac{\varphi(a_{i+1})}{a_{i+1}} < \frac{c}{4m}\cdot\frac{\varphi(a_i)}{a_i} \quad \text{et} \quad a_{i+1} \mid n+i \quad \text{pour } i=0,1,\ldots,k-1.$$

Moyennant les inégalités $\frac{n^2}{2} < \varphi(n)\sigma(n) < n^2$ (cf. [1]), il est facile de déduire des théorèmes analogues concernant la fonction $\sigma(n)$.

Le problème si ces théorèmes sont vrais aussi pour la fonction $\theta(n)$ (nombre des diviseurs de n) reste ouvert.

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MATHEMATICS

On a Certain Problem of K. Kuratowski and A. Mostowski in the Theory of Groups

by

A. EHRENFEUCHT

Presented by K. KURATOWSKI on September 30, 1954

Let C be a set of integers, $c = 2^C$ — the family of its subsets, and $\mathfrak{M}$ — the family of functions $\mu \in C^c$, satisfying the condition

(1) $$\mu(X_1 + X_2) = \mu(X_1) + \mu(X_2) \quad \text{for} \quad X_1 \cdot X_2 = 0, \quad X_1, X_2 \in c.$$

The set $\mathfrak{M}$ forms an Abelian group, the operation being the usual addition of functions, i. e.

(2) $$(\mu = \mu_1 + \mu_2) \equiv (\mu(X) = \mu_1(X) + \mu_2(X)).$$

K. Kuratowski and A. Mostowski have put forward the problem of determining the structure of group $\mathfrak{M}$; they ask in particular whether $\mathfrak{M}$ is isomorphic with group C^c, the addition of functions being defined in the usual way ([1] and problem p. 91, Coll. Mathem. 2, 1951).

The object of the present paper is to confirm the hypothesis of K. Kuratowski and A. Mostowski, i. e. to prove that *group $\mathfrak{M}$ is indeed isomorphic with group C^c.*

Let $\mathfrak{P}$ be the additive group of all sequences $\{x_i\}_{i=0,\pm1,\pm2,\ldots}$ of integers, bounded on both sides (i. e. $\{x_i\} + \{y_i\} = \{x_i + y_i\}$). E. Specker [2] has proved that every subgroup of power $\leqslant \aleph_1$ of group $\mathfrak{P}$ is a free Abelian group. From the continuum hypothesis and from Specker's theorem it follows that:

(3) *Group $\mathfrak{P}$ is a free Abelian group.*

For a given group G we shall denote by H_G the set of all homomorphisms of G into C. H_G constitutes a group, the operation being the usual addition of functions. Obviously, if G is a free Abelian group of power $2^{\aleph_0}$, then group H_G is isomorphic with group C^c. Thus it follows from (3) that

(4) *$H_{\mathfrak{P}}$ is isomorphic with C^c,*

i. e. the proof of the hypothesis of K. Kuratowski and A. Mostowski is reduced to showing that

(5) *groups $H_{\mathfrak{P}}$ and $\mathfrak{M}$ are isomorphic.*

For $\mu \in \mathfrak{M}$ and $\{x_i\} \in \mathfrak{P}$ let us assume that

$$\Phi_\mu(\{x_i\}) = \sum_{m=-\infty}^{m=\infty} m\,\mu\Big(\underset{k}{E}(x_k = m)\Big). \tag{6}$$

It will be noticed that the series on the right is finite since it follows from condition (1) and the boundedness of sequences $\{x_i\} \in \mathfrak{P}$ that

$$\mu\Big(\underset{k}{E}(x_k = m)\Big) \neq 0$$

only for a finite number of $m \in C$.

(7) *For every $\mu \in \mathfrak{M}$ we have $\Phi_\mu \in H_{\mathfrak{P}}$.*

Indeed,

$$\Phi_\mu(\{x_i\}+\{y_i\}) = \sum_m m\mu\Big(\underset{k}{E}(x_k + y_k = m)\Big) = \sum_m m\mu\Big(\sum_l \underset{k}{E}(x_k = l \text{ and } y_k = m-l)\Big)$$

$$= \sum_m m \sum_l \mu\Big(\underset{k}{E}(x_k = l \text{ and } y_k = m-l)\Big) = \sum_m \sum_l [l+(m-l)]\mu\Big(\underset{k}{E}(x_k = l \text{ and } y_k = m-l)\Big)$$

$$= \sum_m \sum_l l\mu\Big(\underset{k}{E}(x_k = l \text{ and } y_k = m-l)\Big) + \sum_m \sum_l (m-l)\mu\Big(\underset{k}{E}(x_k = l \text{ and } y_k = m-l)\Big)$$

$$= \sum_l l\Big(\mu \sum_m \underset{k}{E}(x_k = l \text{ and } y_k = m-l)\Big) + \sum_{m-l} (m-l)\,\mu\Big(\sum_l \Big(\underset{k}{E}(x_k = l \text{ and } y_k = m-l)\Big)\Big)$$

$$= \sum_l l\mu\Big(\underset{k}{E}(x_k = l)\Big) + \sum_{m'} m'\mu\Big(\underset{k}{E}(y_k = m')\Big) = \Phi_\mu(\{x_i\}) + \Phi_\mu(\{y_i\}),$$

$$\Phi_{\mu_1} + \Phi_{\mu_2} = \Phi_{\mu_1+\mu_2} \tag{8}$$

follows from condition (1), which is satisfied by functions $\mu \in \mathfrak{M}$.

(9) *For every $\Phi \in H_{\mathfrak{P}}$ there exists a $\mu \in \mathfrak{M}$ such that $\Phi = \Phi_\mu$.*

For $X \in c$ let $\{X(i)\}$ be the characteristic sequence of the set X, i. e.

$$X(i) = \begin{cases} 1 & \text{if } i \in X \\ 0 & \text{if } i \notin X. \end{cases}$$

Let us assume that $\mu_\Phi(X) = \Phi(\{X(i)\})$.

From $\Phi \in H_{\mathfrak{P}}$ it easily follows that μ_Φ satisfies condition (1), and thus $\mu_\Phi \in \mathfrak{M}$. From definition (6), however, it follows that $\Phi_{\mu_\Phi} = \Phi$, Q. E. D.

(10) If $\Phi_\mu = 0$, then $\mu = 0$.

For if $\mu \neq 0$, e. g. $\mu(X) \neq 0$, then, as follows from (6),

$$\Phi_\mu(\{X(i)\}) = \mu(X) \neq 0,$$

and thus $\Phi_\mu \neq 0$.

From (7), (8), (9) and (10) it follows that Φ_μ is an isomorphism of group $\mathfrak{M}$ with group $H_\mathfrak{p}$; therefore, owing to (5), the hypothesis of K. Kuratowski and A. Mostowski is confirmed.

Institute of Mathematics, Polish Academy of Sciences

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THEORETICAL PHYSICS

Relativistic Motion in a Scalar Field

by

G. MARX and G. SZAMOSI

Presented by L. INFELD on September 21, 1954

The consequences of the relativistic equations of motion are well known in the case of a particle affected by the electromagnetic Lorentz-force. Recently, the force derivable from the scalar potential has also been treated by several authors, and the exact form of the equation of motion has been determined [1], [6]. Starting from the Hamilton-Jacobi equation in a scalar field, J. Werle obtained the following interesting result. Restricting himself to non-relativistic energies, he showed that for purely relativistic reasons the potential, otherwise of attractive character, may lead to the appearence of repulsion. The subject of the present paper is the investigation of this motion in space and time with assumptions more general than the above.

With general assumption the relativistic equation of motion may be written in the form

$$\frac{d}{d\tau}(Mu_i) = F_i, \qquad u_i u_i = -c^2. \tag{1}$$

Here $u_i = dx_i/d\tau$ denotes the four-velocity of the mass point, $icd\tau = \sqrt{dx_r dx_r}$, M the scalar rest mass, and F_i the force vector derivable from the energy-momentum-tensor of the field considered. In general, the equation cannot be satisfied by a constant M, but $M(\tau)$ and the $u_i(\tau)$'s are unknown functions, which can be determined from equation (1). From (1),

$$\frac{dM}{d\tau}u_i + M\frac{du_i}{d\tau} = F_i. \tag{2}$$

Multiplying by u_i we obtain [7]:

$$\frac{dM}{d\tau} = -\frac{1}{c^2}F_i u_i. \tag{3}$$

From this equation it is evident that if the force vector is not perpendicular to the world line of the particle (i. e. to the four-velocity) the rest

mass is varied by the "four effect" of the force and may depend not only on the space co-ordinates of the particle but also on its spin, etc. (In quantum mecanics M is an operator, which generally does not commute with the Hamiltonian). From (3),

$$M(\tau_p) = m - \frac{1}{c^2}\int_{\infty}^{p} F_l dx_l. \tag{4}$$

Introducing (3) into (2) we obtain

$$M\frac{du_l}{d\tau} = \left(\delta_{lk} + \frac{1}{c^2}u_l u_k\right)F_k. \tag{5}$$

We see that the equation of motion can be written in the form: *mass* × *acceleration* in the general case too, with the difference that in this case the force component perpendicular to the world line of the particle must be written on the right hand side [1]. We can separate the force into both a perpendicular and a tangential component:

$$F_l = F_l^{(p)} + F_l^{(t)}, \quad \text{where} \quad F_l^{(p)}u_l = 0, \quad F_l^{(t)} = -\frac{1}{c^2}u_l u_k F_k. \tag{6}$$

Only the perpendicular component has an accelerating effect:

$$M\frac{d^2x_l}{d\tau^2} = F_l^{(p)}. \tag{7}$$

The rôle of the tangential component consists in doing the four-work, which results in varying the rest energy (3):

$$d(Mc^2) = -F_l^{(t)}dx_l. \tag{8}$$

As long as M is positive, the acceleration is parallel to the perpendicular force component. But if the force has a considerable tangential component along a sufficiently long section of the world line, M may become negative. The sign of the acceleration changes during motion also in the case of a constant force. This results, e. g., in preventing the particle from approaching the force centre beyond a certain limit in the case also of a purely attractive force field. For a particle coming from the infinite the acceleration changes its sign at the point P_0 determined by the relation

$$mc^2 = \int_{\infty}^{P_0} F_l\, dx_l. \tag{9}$$

The force $F_l = \frac{e}{c}f_{lk}u_k$ acts on the point charge moving in the electromagnetic field ($f_{lk} = -f_{kl}$). This has no tangential component, therefore the rest mass of the electron does not change during the motion in the electromagnetic field. The case of a nucleon moving in a scalar meson field

is different. Here $F_l = -g\partial_l\varphi$, where $\varphi(x)$ is the attractive potential of the meson field (which becomes more and more negative as the nucleon approaches the force centre, i. e. the nucleus). From (5) we obtain for the rest mass $M = m + g\varphi/c^2$. Taking this into account, the equation of motion (1) assumes the following form:

$$\frac{d}{d\tau}\left[\left(m+\frac{g\varphi}{c^2}\right)u_l\right] = -g\partial_l\varphi. \tag{10}$$

In a static meson field the fourth equation component multiplied by $(-ic)$ expresses the conservation of the energy

$$E = \frac{mc^2+g\varphi}{\sqrt{1-\vec{v}^2/c^2}} = \text{const.} \tag{11}$$

Using this expression the velocity $\vec{v}$ can be eliminated from the first three components of the equation of motion (10), which takes the form

$$m\frac{d\vec{v}}{dt} = -\operatorname{grad} W, \tag{12}$$

where

$$W = \left(\frac{mc^2}{E}\right)^2\left(g\varphi+\frac{g^2\varphi^2}{2mc^2}\right). \tag{13}$$

It seems that the motion of the nucleon is exactly equivalent to a non-relativistic motion under the influence of a potential W. (The *equivalent potential* W in the approximation $E - mc^2 \ll mc^2$ was first derived by Werle from the Hamilton-Jacobi equation of the problem [9]). In the region $g\varphi > -mc^2$ the acceleration is in the same direction as the potential fall: in the region $g\varphi < -mc^2$, however, it is in the *opposite* direction.

The velocity as a function of the space co-ordinates can be determined from the integration of (12) or, in a similar way, from (11)

$$|\vec{v}| = c\sqrt{1-\left(\frac{mc^2+g\varphi(x)}{E}\right)^2}. \tag{14}$$

In a central field the co-ordinates as the function of time can be determined, with the aid of (14), by simple integration, while the process of the motion can already be read from equation (14). While approaching the attractive centre the velocity of the particle increases till it reaches the point r_1 determined by the equation $g\varphi(r_1) = -mc^2$. At this point (at the boundary of the repulsive zone) the velocity of the nucleon reaches that of light and its acceleration becomes zero. Reaching the velocity of light presents no difficulty because the energy and the total mass remain finite owing to the fact that the rest mass simultaneously becomes zero. Entering the region $g\varphi < -mc^2$ the velocity of the particle begins to decrease; in the case of radial motion the velocity becomes zero at the distance r_2 determined by the equation $g\varphi(r_2) = -mc^2 + E$. Here the motion of the nu-

cleon turns towards the opposite direction, the particle speeds up again till it attains the velocity of light, then, leaving the repulsive zone, it slows down and passes into the infinite. It is worth noting that the boundary of the repulsive zone does not depend on the initial conditions and the possible presence of an electromagnetic field.

If the meson field originates from a nucleon considered as fixed $\varphi = -\frac{g e^{-\mu r}}{r}$, the boundary of the repulsive zone is at the distance $2r_0$ determined by the equation

$$g^2 \frac{e^{-\mu(2r_0)}}{(2r_0)} = mc^2. \tag{15}$$

On the analogy of the classical electron radius r_e, defined by the equation $e^2/2r_e = m_e c^2$, it seems useful to call r_0 the *classical nucleon radius* (r_0 is smaller than the range μ^{-1} of the nuclear force, its value is of the order of 10^{-14} cm.). If a nucleon approaches another to within a distance corresponding to the double nuclear radius, the attraction between them is converted into repulsion. That is why, in the case of non-relativistic initial velocities, the nucleons can never approach one another so as to be within distances much smaller than r_0. Our result can be further illustrated by saying that the relativistic treatement attributes a "finite size" to the nucleon originally assumed to be point-like, and its size is characterized by the classical nucleon radius r_0. As long as the nucleons do not come into "contact" they attract each other, but as soon as they are in "contact", repulsion takes place between them, which soon results in the "recoil" of the "colliding" nucleons. In the repulsive region and in its neighbourhood $e^{-\mu r} \simeq 1$. Taking this into account the equation of motion can easily be integrated. For radial motion the relation between the distance r and the time t is

$$t = t(r_0) + \frac{r_0}{c}\left[\left(\frac{r}{r_0} - 1\right)^{1/2} + \frac{1}{3}\left(\frac{r}{r_0} - 1\right)^{3/2}\right]. \tag{16}$$

It can be seen that the stay of the nucleon in the repulsive region is very short; it is of the order of 10^{-24} sec.

If a nucleon moves in a field established by several other nucleons, the potential acting on it is $\varphi(\vec{r}) = -\sum_l g e^{-\mu|\vec{r}-\vec{r}_l|}/|\vec{r}-\vec{r}_l|$, from which, according to (14), the equivalent potential determining the motion of the nucleon is

$$W = \sum_l \left(-g^2 \frac{e^{-\mu|\vec{r}-\vec{r}_l|}}{|\vec{r}-\vec{r}_l|} + \frac{g^4}{2mc^2} \cdot \frac{e^{-2\mu|\vec{r}-\vec{r}_l|}}{|\vec{r}-\vec{r}_l|^2}\right) + \sum_{i \neq j}' \frac{g^4}{mc^2} \cdot \frac{e^{-\mu|\vec{r}-\vec{r}_i|}}{|\vec{r}-\vec{r}_i|} \cdot \frac{e^{-\mu|\vec{r}-\vec{r}_j|}}{|\vec{r}-\vec{r}_j|}. \tag{17}$$

(We have assumed that $E \simeq mc^2$). It can be seen that the motion taking place in the field of several nucleons (e. g. of a nucleus) is equivalent to a non-relativistic motion in which, in addition to the two-body forces,

three-body forces also act. The attractive force between two nucleons is decreased by the presence of further nucleons. The same applies to the case of extended forces.

The classical motion of nucleons must be treated exactly as a many-body problem, taking into account radiation phenomena. This is, however, a difficult problem which so far has remained unsolved, and which requires further investigation. The same applies to the problem of motion in a pseudoscalar meson field. In this case too the rest mass is not constant and, consequently, similar phenomena must also appear.

A more detailed account of the present calculation will appear in Acta Physica Hungarica.

Our thanks are due to Docent J. Werle and Docent J. Plebański for interesting discussions which initiated the above investigations.

Physical Institute of the Roland Eötvös University, Budapest

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THEORETICAL PHYSICS

Order of Magnitude of the Field Mass of a Nucleon in a Non-linear Meson Field Theory

by

M. MIĄSEK and A. LEGATOWICZ

Presented by L. INFELD on September 23, 1954

One of the fundamental difficulties in linear field theories, in both electrodynamics and mesodynamics, arises from the fact that the energy of a field of elementary particles is infinite, when they are treated as point sources of the field.

In electrodynamics the non-linear theory of Born and Infeld [1], [2], introducing the Lagrangian in a modified form, removes this difficulty. The question arises as to whether in mesodynamics analogous results may be obtained by using the Lagrangian of non-linear electrodynamics generalized by introducing the Proca-term for meson fields.

This problem has already been treated quantitatively by Walsh [4]. The purpose of the present paper is to give a more detailed and, at the same time, a more universal analysis of the results obtained when a Lagrangian thus modified is used in the case of a static scalar-field with spherical symmetry corresponding to one nucleon.

We are considering a class of non-linear theories fulfilling the following conditions:

I. The field equations become, for weak fields, equations of linear mesodynamics.

II. The density of the field energy is everywhere finite and has the greatest value in the centre of the particle.

The Lagrangian of a field fulfilling the conditions given above will have the form:

$$L = b_m^2 F(b_m, \psi^2, \psi'^2), \tag{1}$$

and we shall have

$$L \to \begin{cases} -\frac{1}{2}(\psi'^2 + K^2\psi^2) = L_y \text{ , for } \psi \to 0,\ \psi' \to 0,\ K = \frac{m_\pi c}{\hbar} \\ -b_m^2 = L_{\max} \quad \text{for} \quad r \to 0. \end{cases} \tag{2}$$

The field energy of a particle will be expressed by the formula:

$$E = \int_0^\infty -Lr^2\,dr. \tag{3}$$

The field equations may be obtained from the variational principle

$$\delta \int_0^\infty Lr^2\,dr = 0 \tag{4}$$

with the assumption, that for $r \to \infty$ the field vanishes.

It is well known that the solution of the field equation corresponding to the Lagrangian L_y is the Yukawa-potential

$$\psi_y = g\frac{e^{-Kr}}{r}. \tag{5}$$

In our case the potential (5) represents the asymptotic form of the solution of the field equation for $r \to \infty$.

To get an approximate estimation of the field energy we assume that potential (5) is a satisfactory approximation of the solution of the field equation in the region where $r > r_0$ and where for $r = r_0$ the following condition is fulfilled:

$$\left|\frac{L(r_0) - L_y(r_0)}{L(r_0)}\right| = \lambda < 1. \tag{6}$$

Introducing potential (5) into equation (6) we are able to calculate for every form of the Lagrangian (1) the quantity

$$b_m = f(g, r_0, \lambda). \tag{7}$$

To estimate the upper limit of the field energy we use formula (3) assuming for $r > r_0$ the potential (5) and for $0 < r < r_0$,

$$L = -b_m^2 = L_{\max}. \tag{8}$$

To estimate the lower limit of the field energy we proceed analogously assuming for $0 < r < r_0$,

$$L = L_y(r_0) = L_{\min}. \tag{9}$$

The approximate value of the field energy may be calculated using formula (3) and assuming that in the region $0 < r < r_0$ we have

$$L = L_{\max} - \frac{L_{\max} - L_{\min}}{r_0}\,r. \tag{10}$$

When we take, as a particular example, the Lagrangian in the form

$$L = b_m^2\left\{\sqrt{1 - \frac{1}{b_m^2}(\psi'^2 + K^2\psi^2)} - 1\right\}, \tag{11}$$

we get from the variational principle (4) the following field equation

$$\varphi'' = \varphi - 2\frac{\varphi'}{x} - 2\frac{\varphi'^2}{1-\varphi^2}\left(\varphi - \frac{\varphi'}{x}\right), \tag{12}$$

where

$$\varphi = \frac{K}{b_m}\psi \tag{13}$$

$$x = Kr. \tag{14}$$

On the basis of a detailed analysis of equation (12) it may be shown that:

1. There exists a solution of equation (12) which has for $x \to \infty$ the asymptotic form:

$$\varphi_\infty = A\frac{e^{-x}}{x}. \tag{15}$$

2. The function $\varphi_0 = \sin(x+\alpha)$, where α is an arbitrary constant, is a singular solution of equation (12).

3. The solution of equation (12) which for $x \to \infty$ assumes form (15) is a monotone decreasing function.

4. The solution of equation (12) which for $r \to \infty$ assumes form (15) has, for $x \to 0$, a contact of the fourth order with the function $\sin(x+\alpha)$, for a particular value of the parameter α, which is a function of A.

Hence it follows that conditions I and II given at the beginning are fulfilled by the Lagrangian (11).

So, using our formulas for the estimation of the field energy of a particle, we get from (6), (11), (3), (8) and (9),

$$b_m^2 = \frac{1}{2}\frac{L_y(r_0)}{\lambda(\lambda-1)}$$

$$E_{\max} = \frac{1}{2}g^2K\frac{1}{e^{2x_0}x_0}\left\{1 + x_0 + \frac{1}{6}\frac{1}{(1-\lambda)\lambda}(2x_0^2 + 2x_0 + 1)\right\} \tag{17}$$

$$E_{\min} = \frac{1}{2}g^2K\frac{1}{e^{2x_0}x_0}\left\{1 + x_0 + \frac{1}{3}(2x_0^2 + 2x_0 + 1)\right\}. \tag{18}$$

We obtained the smallest difference between $E_{\max}$ and $E_{\min}$ assuming $\lambda = 0{,}5$. The energy calculated for this value of the parameter λ is expressed by

$$E = 5.463 \cdot 10^{-4}\frac{g^2}{x_0 e^{2x_0}}\left\{1 + x_0 + \frac{3 \pm 1}{6}(2x_0^2 + 2x_0 + 1)\right\} Mc^2, \tag{19}$$

where the charge of an electron is taken as a charge unit and the rest mass of a nucleon as an energy unit.

The approximate value of the energy calculated using formula (10) will be

$$\tilde{E} = 5.463 \cdot 10^{-4}\frac{g^2}{x_0 e^{2x_0}}\left\{1 + x_0 + \frac{2.5}{6}(2x_0^2 + 2x_0 + 1)\right\} Mc^2. \tag{20}$$

According to our condition I, that for weak fields the theory becomes a linear one, we assume in numerical calculations for the parameter g the value $g = 5e$, which, for linear theory, is experimentally justified.

The calculations were made for two cases:

A. assuming r_0, i. e. the range of non-linear modification of the field, to be of the order 10^{-13} cm., which is in agreement with the experimental data for the dimensions of a nucleon;

B. assuming that the whole mass of the nucleon is the field mass.

In case A, assuming $g = 5e$, $x_0 = 1$, $r_0 = 1.4 \cdot 10^{-13}$ cm., we obtained the following values for the field energy of the particle:

$$E_{\max} = 9.8 \cdot 10^{-3} = \frac{1}{101} Mc^2$$

$$\tilde{E} = 7.5 \cdot 10^{-3} = \frac{1}{133} Mc^2$$

$$E_{\min} = 6.8 \cdot 10^{-3} = \frac{1}{147} Mc^2$$

In case B, assuming $g = 5e$, $E = 1 Mc^2$, we calculated the range of the non-linear modification of the field as follows:

$$r_0 \max = 3.14 \cdot 10^{-15} \text{ cm.}$$
$$\tilde{r}_0 = 2.69 \cdot 10^{-15} \text{ cm.}$$
$$r_0 \min = 2.52 \cdot 10^{-15} \text{ cm.}$$

The results given above are in agreement with those obtained in another way by Walsh. They show that it is wrong to expect that a non-linear generalization will be able to explain the whole mass of the nucleon as a field mass connected with the charge of a meson field.

Indeed, in case A, assuming reasonable values for g and r_0 we see that for a large class of non-linear theories the field mass amounts only to obout 1/137 of the whole mass of the nucleon.

And in case B, if it is assumed that the whole mass of the nucleon is the field mass, it follows that r_0 should be of the order 10^{-15} cm., which is not in agreement with experimental data for the dimensions of a nucleon.

We should like to express our sincere thanks to Professor L. Infeld and to Docent J. Plebański for their valuable advice during the preparation of this work.

Institute of Theoretical Physics, University of Warsaw

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CHEMISTRY

The Electric Conductivity of Crystalline Methylene Blue

by

K. GUMIŃSKI and W. ROMANOWSKI

Presented by M. ŚMIAŁOWSKI on July 4, 1954

Until recently, practically only inorganic substances were included in the group of bodies termed semi-conductors. The phenomena of electric conductivity, photo-conductivity, thermo-electric and galvanomagnetic effects in these substances were explained according to the band theory of crystals.

Organic substances, on the other hand, were classed among insulators, as distinguished by a considerably higher resistivity and a closer though also exponential dependence of their resistivity upon temperature. However, research in this direction was not pursued in spite of the fact that certain data as to the external [1] and internal [2] photo-electric effects have long been known. It was not until recently that certain sporadic works, particularly those of the Soviet scientist A. T. Vartanian [3] and some Japanese researchers [4], on the subject of the semi-conductive properties of organic substances began to appear.

Research in this direction has also been undertaken by the Department of Physical Chemistry of the Institute of Technology in Wrocław, and the first positive results have been obtained with a thiazine dye-methylene blue [5].

Apparatus and Method of Measurement

When testing the electric resistance of organic substances and its dependence upon temperature, allowance must be made for the fact that the resistivity will be in general much higher than in the case of inorganic substances, and the temperature range in which the sample can be tested will be more restricted. This is either because these substances usually possess a low melting point, or because they decompose at higher temperatures. Since these resistances are very high, it is advisable to use electrostatic methods of measurement.

In our investigations, the resistance of samples was measured by the condenser discharge method. The voltage of a charged condenser of a ca-

pacity C, discharged by the resistance R, will drop according to the law:

$$V = Vo e^{-\frac{t}{RC}},$$

where $Vo =$ the voltage at the moment t_0, $V =$ the voltage at the moment t. By plotting the successive temporary values $\ln \frac{V_0}{V}$ against the time t, we shall obtain a straight line. The tangent of the angle of inclination of this line will amount to $\frac{1}{RC}$. We can now, knowing the capacity C of the condenser, estimate the value of R or $\frac{1}{R}$. The scheme of arrangement is represented in Fig. 1.

A Wulf type quadrant electrometer connected heterostatically was used for measuring the voltage. Condensers with a capacity of 420 and 16250 pF were employed. The capacity of the electrometer and conductors amounted to approximately 20 pF. The condenser was charged from an anode battery up to 120 V.

The details of experimental procedure were as follows. A $20 \times 30 \times 1.5$ mm. quartz plate was thoroughly cleaned, and two rectangular electrodes, spaced roughly 1 mm. apart, were formed of silver by vaporization through a suitable template in a high vacuum. These electrodes were gold-plated to ensure superior durability. A few drops of a concentrated aqueous solution of methylene blue were then introduced into the gap between the electrodes and quickly evaporated at a temperature of c. 100° C. in an electric drying apparatus. In this way a crystalline layer of the dye was obtained between the electrodes. The compactness of this layer was examined under a microscope. The quartz plate, with the elec-

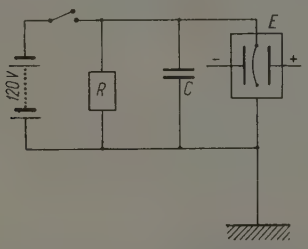

Fig. 1. Scheme of arrangement of measurements

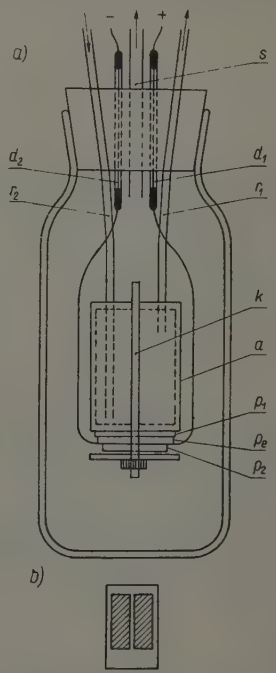

Fig. 2. a) apparatus for testing the resistance of the layers, b) plate with electrodes

trodes so prepared and with the layer of dye, was then placed in a suitable vessel immersed in an oil bath. The temperature of the latter was brought up to c. 110° C., and the vessel evacuated to a high vacuum. This was done in order to remove any residue of the solvent and of water adsorbed on the surface, and usually took 2 hours. The sample was then quickly placed in the apparatus illustrated in Fig. 2.

This device consists of a cylindrical metal vessel a, in which circulates the liquid from a thermostat provided with supply pipes r_1 and r_2 for the liquid, and a clip k with screws to enable the plate holding the electrodes and the insulators to be connected to the vessel. The plate p_e holding the electrodes is insulated from the vessel and clip by means of thin quartz plates, p_1 and p_2. The vessel is inserted into the rubber stopper which seals the outer glass vessel. Quartz tubes containing the leads d_1 and d_2 to the electrodes and a glass tube s, which forms the connection between the interior of the vessel and the vacuum pump, are passed through the rubber stopper. To remove all traces of steam from the vessel, a small quantity of silica gel was placed on the bottom. The efficacy of the insulation was checked prior to every measurement by placing the plate carrying the electrodes in the device and testing its resistance before depositing the layer of dye.

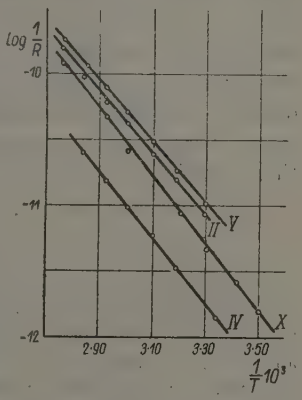

Fig. 3. Dependence of $\log \frac{1}{R}$ upon $\frac{1}{T}$ for crystalline layers of methylene blue. Roman numerals denote the individual samples.

After the sample had been introduced, the vessel was evacuated to about 1 mm. Hg, and the metal vessel connected with a thermostat set for the temperature required.

Measurements were carried out, starting from room temperature and continuing under progressively increasing temperatures. After reaching the maximum temperature possible in the experimental conditions (90°), measurements were carried out at decreasing temperatures. The resistance of the samples at room temperature was of the order of 10^{11} ohms, and at a temperature of 90° C., of the order of 10^9 ohms.

Results

We have by these means arrived at a temperature relation of the resistance of the solid dye ranging from 10° to 90° C. All samples tested, the number of which amounted to twenty, revealed an identical character of relationship. The results of measurements for a number of samples picked at random are shown in the graph represented in Fig. 3. The mean activation

energy computed from the total of all samples investigated, according to the formula $\sigma = \sigma_0 e^{-E/2KT}$, amounted to 0.92 ± 0.05 eV.

These results were obtained for pure methylene blue, free from zinc chloride. ($ZnCl_2$ appears as an impurity in methylene blue, since it is used in the preparation of this dye). The dyes investigated were made by E. Merck of Darmstadt and Aniline Division N. Y. Samples of pure methylene blue as made by Hoechst-Lucius Brüning gave activation energy values of 1.12 and 1.17 eV.

Tests were also carried out on methylene blue as used in dye works in the form of the double salt containing zinc chloride. The resistance of these samples, prepared in an identical manner, was roughly 100 times higher and their activation energy substantially higher within the temperature range specified above. Similar values for resistance and activation energy (1.6 eV) were obtained for a pure Merck preparation after a considerable quantity of zinc sulphate had been added.

The values for E obtained for the pure dye were slightly less than those obtained by A. T. Vartanian [3].

Summary

The dependence of electric conductivity upon temperature was investigated in the case of methylene blue in the form of thin crystalline layers within the range from 10° to 90° C. The conductivity found seems to have the character of a semi-conductor, with an activation energy of approximately 0.92 eV. Methylene blue containing impurities in the form of zinc chloride or zinc sulphate revealed, at room temperature, a resistance higher by two orders than that of the pure dye, whereas the activation energy increased to c. 1.6 eV.

Laboratory of Physical Chemistry, Institute of Technology, Wrocław
Institute of Theoretical Chemistry, Jagellonian University, Cracow

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CHEMISTRY

Suggested Standard for Low Dielectric Constant

by

J. HURWIC

Presented by M. ŚMIAŁOWSKI on July 8, 1954

Measurement, for various purposes, of the dielectric constant, involves the necessity of gauging the condenser vessel by means of a liquid having a dielectric constant amounting to about 2.

It is essential that the standard be closely defined, accurately reproducible and readily adaptable for use.

Benzene, generally used for this purpose and having a dielectric constant of only slightly more than 2, does not meet the first two requirements, for the following reason:

The dielectric constant of water amounts to roughly 80; hence, a very small proportion of water added tends to increase the dielectric constant. Experiments carried out by Schupp and Mecke [1] reveal that as little as 4.2 mg. (0.00023 ths of a mol) of water added per litre of benzene cause the value of the dielectric constant to vary by one unit in the fourth decimal digit. It is for this reason that benzene must be carefully dehydrated (say, by distillation over sodium and storage over sodium) and every care taken to prevent access of moisture during the test.

This is a serious handicap in tests and involves the use of complicated apparatus. The maintenance of benzene, in spite of it having been carefully dehydrated, in an anhydrous state is, moreover, in practice, extremely difficult. A certain quantity of water vapour from air is bound to penetrate during the test procedure into the liquid, and obviously air is never perfectly dry. This quantity of water vapour is very small, but sufficient to increase the dielectric constant. Contact between 10 cc. of air of normal humidity at room temperature with 20 ml. of dehydrated benzene causes a perceptible increment in the dielectric constant [1]. Worse still, the quantity of moisture absorbed varies in connection with moisture content in the air, the time benzene remains in contact with air, and other factors.

Water dissolves in benzene in minute quantities; according to Rosenbaum and Walton [2] who investigated, by means of calcium hydride,

the degree of solubility of water in certain organic solvents, the quantity of water dissolved at 20° C. in 100 g. of benzene amounts to 0.0573 g. Although water added to benzene does affect the value of the dielectric constant, yet it does not change its value significantly.

The circumstances referred to have prompted me to suggest a saturated solution of water in benzene as a standard for a low dielectric constant. Such a solution can be prepared by intimately mixing benzene with distilled water in a vessel placed in a mechanical shaker. The dielectric constant of the solution thus obtained amounts, at the temperature of 20° C., to 2.2942 [1].

The solution mentioned meets all requirements which such a standard is expected to fulfil.

This solution is successfully used by me as a solvent in the determination of dipole moments [3], [4].

Department of Physical Chemistry, Institute of Technology, Warsaw.

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CHEMISTRY

Polyazeotropic Mixtures Containing Two or More Series of Homologues. II

by

W. ŚWIĘTOSŁAWSKI

Communicated at the meeting of December 13, 1954

1. Classification of Polyazeotropic Mixtures

In the preceding paper of this series [1] coal tar and tar oils were called polyazeotropic mixtures. In this paper an attempt is made to classify the polyazeotropic mixtures more precisely. Namely, if a liquid organic raw material or any product obtained by its fractional distillation contains two series of homologues and their isomers, for instance, if one is composed of paraffins (H), the other of aromatics (Ar), it is reasonable to call it a two-series polyazeotropic mixture. Consequently, for liquids containing three or more series of homologues and their isomers, the term polyazeotropic mixture should be supplemented by the number of the series of homologues and their isomers which are the constituents of that mixture.

Coal tar or at least some tar oils contain not less than six series of homologues, namely: (H), (Ar_1), (Ar_2), (F), (P), (Am). The symbols represent: (H) — paraffins, (Ar_1) — one-ring aromatics, (Ar_2) — two-ring aromatics, (F) — phenols, (P) — pyridine bases, (Am) — aromatic amines. We purposely omit the series of olefines (O) and the naphthenes (N), because they are found in high temperature coal tar in small quantities and, in addition, their azeotropic ranges [2] differ little in respect to the paraffins and aromatics so that they do not exert any notable influence on the course of distillation. In low temperature coal tar, however, the concentrations of olefines are considerably larger, and their influence on distillation cannot be neglected.

2 Polyazeotropic Mixtures Composed of Two Series of Homologues

Let us suppose, for simplification, that large quantities of two mixtures (H) and (Ar) have been prepared by mixing equal molar quantities of each of the representatives of series (H) on the one hand and of series (Ar)

on the other. Using these two mixtures we would be able, for instance, to prepare the following three polyazeotropic mixtures in which m changes from 19 to 1:

Number	Weight % of H	Weight % of Ar	$C_{H_l} = mC_{Ar_j}$	m
1	95%	5%	$C_{H_l} = 19C_{Ar_j}$	19
2	75%	25%	$C_{H_l} = 3C_{Ar_j}$	3
3	50%	50%	$C_{H_l} = C_{Ar_j}$	1

Suppose that the azeotropic ranges [2] of all the components of series (Ar) are known; for instance, for Ar_j we may write:

$$Z_{Ar_j}(H) = t_{Hk} - T_{H_e},$$

where H_k and H_e are the representatives which form with Ar_j two tangent azeotropes [3]. Suppose also that the boiling temperatures t_{H_l} and t_{Ar_j} are equal or almost equal to each other.

Let us consider the phenomena which would take place if the three mixtures mentioned above were submitted to a fractional distillation. Because of the small amounts of each of the members of series (Ar) in mixture 1, the component Ar_j would form successively an almost tangent zeotropic isobar with the representative H_{e-1}, a tangent one with H_e and an almost tangent azeotropic isobar with H_{e+1}. When distilling mixture 2, we would observe that the corresponding azeotropes (Ar_j, H_{e+2}), (Ar_j, H_{e+3}) would be formed. Finally, if a mixture of equal concentrations of the representatives of both series were submitted to fractional distillation, curve III shown in Fig. 1 *c* would represent graphically the large increase in concentration of the component Ar_j.

We have examined the case in which the differences in boiling temperatures of components $t_{H_{i-1}}, t_{H_i}, t_{H_{i+1}}$ and $t_{Ar_{j-1}}, t_{Ar_j}\, t_{Ar_{j+1}}$ were so large that no superimposing of concentration curves took place. If these differences were smaller, azeotropes formed by Ar_{j-1} and Ar_{j+1} would be admixed to those formed by the component Ar_j. These phenomena are frequently encountered in cases of industrial batch distillations of different polyazeotropic mixtures.

Let us, however, return to our simplified case and formulate some of the phenomena taking place when a two-series polyazeotropic mixture is submitted to fractional distillation. First, if the molar concentrations of each representative of series (Ar) are smaller than those of the components of series (H), such a polyazeotropic system may be called a two-series (H) — polyazeotropic mixture. Symbol (H) denotes the preponderance of concentrations of components (H) as compared with those of (Ar). In this case any substance Ar_j undergoes distillation within a narrow range of tempe-

ratures close to the lowest one, t_{H_e}, at which Ar_j forms with H_e a tangent isobar with respect to the horizontal line drawn through point t_{H_e}. With an increase of the molar concentration of Ar_j, there is a corresponding increase in the number of azeotropes formed with the representatives of series (H). Secondly, if the concentrations of any of the components of both the (H) and (Ar) series, for instance, H_i and Ar_j, boiling practically at the same temperatures, become equal ($m=1$), the preponderance of concentrations of the components of any of these series disappears. In that case the largest amount of azeotrope (Ar_j, H_i) is found in the receiver. At the moment of maximum concentration of Ar_j in the distillate the condensation temperature is close to 0.5 ($t_{Ar_j}-t_{H_e}$). Similar phenomena take place, if the polyazeotropic mixture is prepared so that the components of the series (Ar) are in larger concentrations than those belonging to series (H). In the latter case curves *I*, *II* and *III* refer to component H_i and not to Ar_j.

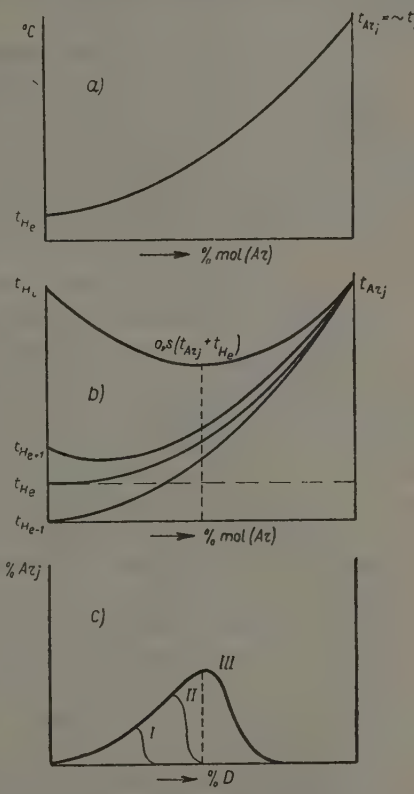

Fig 1. A section of the distilling curve is represented in scheme *a*. It starts with condensation temperature t_{H_e} and ends with t_{Ar_j}. Scheme *b* shows the shape of several boiling temperature isobars starting with the almost tangent "zeotropic" one, $t_{H_{e-1}}t_{Ar_j}$, and ending with $t_{H_{e+1}}t_{Ar_j}$, which is characterized by a relatively small depression Δt. In scheme *c* curves *I*, *II* and *III* correspond to the changes in concentrations of Ar_j in the distillates, when mixtures 1, 2 and 3 are submitted to fractional distillation

When dealing with industrial products, we have to take into consideration the fact that they are more complicated polyazeotropic mixtures than those mentioned above. Some similarity, however, may be noticed if we examine the distillation of low, medium and high boiling gasolines. In the case of low boiling gasoline, benzene and then toluene start to appear in the receiver at lower temperatures than their own. The larger the lower portion of the azeotropic range [2] of Ar_j, the lower is the boiling temperature of the mixture containing a small amount of the corresponding aromatic hydrocarbon. It is a well-known fact that benzene and toluene

may be easily entrained by nonaromatic hydrocarbons which form with benzene or toluene tangent zeotropes and azeotropes before the next aromatic hydrocarbon starts to appear at the top of the distilling column. On the other hand, the xylene isomers boil so close to one another that curves *I*, *II* and *III* shown in Fig. 1c become superimposed one on top of the other.

For the sake of simplicity we do not mention that gasoline contains naphthenes as the third series of homologues. They do not produce any notable influence on the shape of the distilling curve. They form azeotropes with aromatic hydrocarbons (Ar_j, N_l), which behave in a similar way to those composed of H_l instead of N_l, where symbol N stands for any representative of the naphthene series.

3. Methods for Studying More Complicated Polyazeotropic Mixtures

Much more complicated phenomena take place if more than two series of homologues are present in a polyazeotropic mixture. Beside low and high temperature coal tar and different tar oils, other industrially important liquids belong to this type of polyazeotropic mixture. A special technique based often on fractional distillation and on ebulliometric investigation of closely boiling fractions collected in the course of a batch distillation carried out on a pilot plant or industrial column, has to be applied in order to obtain more information concerning the phenomena associated with both an azeotropic and a zeotropic distillation of these mixtures. By removing phenols, organic bases, olefines, etc. it becomes possible to reduce the number of homologous series. In that way a direct comparison of the distillation curves obtained before and after the reduction of the number of homologous series, offers important information concerning the influence exerted by one, two or sometimes three series of homologues and their isomers [3].

In some particular cases certain components may be removed by crystallization and the shape of the distilling curves before and after the removal of those components may also be directly compared [3], [4].

Another method consists in adding a large excess of a certain component so as to produce all the azeotropes formed by it. The result of this procedure is often important for understanding some phenomena taking place during the distillation [5].

A group of Polish scientists are at present engaged in carrying out experiments in order to find the most suitable conditions for studying different polyazeotropic mixtures.

Summary

1. The polyazeotropic mixtures have been classified according to the number of homologous series contained in them.

2. In order to examine the most typical phenomena taking place in the course of a batch distillation, it was assumed that two liquid systems had been prepared, one (H) containing equimolecular quantities of representatives of one homologous series, another

(Ar)-equimolecular quantities of the other series. Afterwards, typical mixtures were prepared by mixing these two systems so as to obtain mixtures with varying ratios $C_{H_i} : C_{Ar_j} = m$.

3. If ratio m is large enough, each of the representatives of series (Ar) is collected in the receiver at temperatures differing slightly from t_{H_e} of the component H_e, which forms with Ar_j a tangent azeotrope.

4. For studying the distillation phenomena taking place during the batch distillation of a polyazeotropic mixture, the distillation and ebulliometric methods are combined with the removal of acid, basic and neutral components. Sometimes a certain component is removed by crystallization. In other cases a large amount of an agent is added, which is able to entrain azeotropically a large number of components belonging to one, two, or sometimes three series of homologues.

Department of Physical Chemistry, University of Warsaw

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CHEMISTRY

A Method for Determining the Composition of Quaternary Heteroazeotropes

by

W. ŚWIĘTOSŁAWSKI and A. GALSKA

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1. Introductory Remarks

In 1951 K. Zięborak proved the existence of quaternary positive azeotropes of the type (B, E, W, H), where B stands for benzene, E — for ethanol, W — for water and H — for one of the hydrocarbons of the paraffins or naphthenes boiling at temperatures ranging from 68° to 100° C. In his experiments both the ebulliometric and distillation methods were used. [1]. The first experiment consisted in examining the shape of the boiling and condensation isobars obtained for mixtures of two ternary positive heteroazeotropes: (B, E, W) and (H_i, E, W). Symbol H_i represents one of the three hydrocarbons: 2,2,4 pentane, *n*-heptane and cyclohexane. If minimum boiling and condensation temperatures were found, it was concluded that a quaternary azeotrope did exist. It must be borne in mind, however, that the composition of the mixture boiling and condensing at a minimum temperature may, or may not, correspond to the composition of the quaternary azeotrope under examination.

For years different investigators have used the distillation method for preparing azeotropes. Since, however, the corresponding isobars are usually very flat in the region close to the azeotropic point, especially if ternary and quaternary azeotropes are involved, some doubt may be entertained as to whether the main fraction collected in course of a fractional distillation will really have the composition of the azeotrope examined. For this reason Zięborak and Galska and also the authors of this paper have tried to find other methods for determining more exactly the composition of quaternary positive heteroazeotropes. One of these methods is described in this paper.

2. Ebulliometric Examination of Quaternary Azeotropes

Let us assume that a mixture of four components, B, E, W and C, the last being cyclohexane, has been submitted to a fractional distillation and that the main fraction has been collected in a sufficient quantity for car-

rying out four independent ebulliometric measurements. After filling a differential ebulliometer with a corresponding amount [2] of the main fraction, we have to determine the difference between the boiling and condensation temperatures of the liquid examined in order to determine the ebulliometric degree of purity of that liquid which is considered to be close to the composition of the quaternary heteroazeotrope. After this measurement has been completed we start to determine the shape of sections of the boiling and condensation isobars by introducing into the ebulliometer, several times in succession, small amounts of one of the components, e. g. water. Two different eventualities may occur: either the boiling and condensation temperatures may show a gradual increase with the increase in the amount of water added, or minima of both boiling and condensation temperatures will appear. If the latter occurs, it is a proof that the mixture examined contains less water than that which corresponds to the composition of the azeotrope under examination.

Afterwards, we have to examine the shape of three other sections of the isobars by adding, each time to a new portion of the main fraction, other constituents of the azeotrope. In this way we obtain results which enable us to draw a diagram similar to that shown in Fig. 1.

In the case examined, one isobar only was characterized by minimum boiling and condensation temperatures. It was obtained by adding water to the main fraction $F(I)$. In addition, it is easy to see that isobars I and II show a relatively rapid increase in boiling temperatures with an increase in the amount of the component added. On the other hand, when benzene or cyclohexane were added to the main fraction the two isobars III and IV merged along a portion of the concentration axis, as shown in Fig. 1. It should be emphasized that both benzene and cyclohexane were found almost exclusively in the upper phase.

3. Conclusions

The following conclusions may by drawn from the experiments just described: if minimum boiling and condensation temperatures are found on a corresponding isobar, it proves that the main fraction F contains less of that component the addition of which is associated with the appearance of a minimum. No more than three isobars may contain a minimum of the boiling temperature. In order to obtain by fractional distillation a composition identical with or very similar to that of the quaternary azeotrope, it is necessary to use an excess of those components which form isobars sharply inclined to the concentration axis, (see curves I and II in Fig. 1). On the other hand, the two remaining components, i. e. those giving curves III and IV, should be used in small amounts. The concentration ratio of these two components should be equal or almost equal to that which characterizes them in the azeotrope.

As to the composition of the quaternary azeotrope consisting of ben-

zene, ethanol, water and cyclohexane, the following propositions seem to be the most probable: 54.3% (54.0) of cyclohexane, 19.4% (17.4) of ethanol, 6.1% (7.1) of water and 20.4% (21.5) of benzene. The value found by Zięborak [1] by the distillation method are given in brackets.

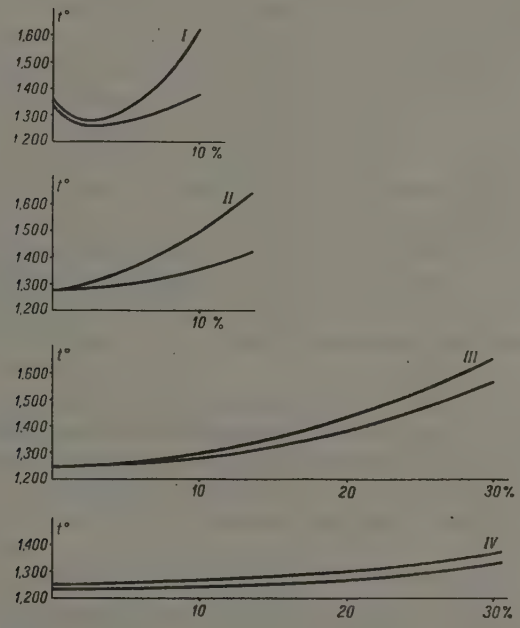

Fig. 1. Four sections of boiling and condensation temperature isobars obtained by examining the changes in the boiling temperatures of the main fraction *F*, when adding: water *I*, ethanol *II*, benzene *III* and cyclohexane *IV*. In order to determine the shapes of the isobars, the differential ebulliometer was filled each time with some amount of the main fraction (*F*)

4. Final Remarks

The conclusions mentioned above were submitted to experimental verification. Namely, the main fraction collected into the differential ebulliometer was characterized by a difference $\Delta t = 0.010^0$ C. between the boiling and condensation temperatures. Therefore the distillation of an excess of *W* and *E* together with small amounts of *B* and *C* gave proper results.

One more remark must be added. In spite of the good results obtained by using the ebulliometric method for examining the composition of the quaternary, and any other azeotrope, both the distillation and ebulliometric methods have to be applied for a proper examination of the composition and the boiling temperature of the azeotrope.

The main advantage of the ebulliometric method is that it permits direct measuring of the boiling temperatures of mixtures until a minimum boiling temperature is observed. Both the boiling and the condensation temperatures may be determined, so that direct control of the purity of the initial material, as well as of the azeotrope obtained, may be maintained by applying the usual ebulliometric measurements [2].

The distillation method for the preparation of an azeotrope has another advantage, namely, that in most of the experiments the impurities present in the initial mixture, after the latter has been submitted to careful fractional distillation, are found usually either in the forerunnings or in the bottom product. Owing to this fact the distillate collected within a very narrow temperature range is practically composed of pure components. The composition, however, of the main fraction may be shifted somewhat towards one side or the other of the boiling temperature isobar so that the main fraction may not correspond exactly to the composition of the azeotrope examined. For this reason, the method described in this paper for determining the shape of four — or three in the case of a ternary azeotrope — boiling temperature isobars, is capable of providing a definite answer to the question whether or not the main fraction corresponds to the composition of the azeotrope. In addition, the ebulliometric method is the only one which makes it possible to establish by direct measurements the shape of the boiling and condensation temperature surfaces corresponding to these two isobars.

Summary

1. A combined distillation and ebulliometric method for examining quaternary azeotropes was described.

2. The boiling and condensation temperature isobars of mixtures of the main fraction — the latter having a composition close to that of the azeotrope under examination and the quantities of each of the components of the azeotrope being known — have to be determined so as to prove whether or not there is a lack or an excess of each of the components in the main fraction.

3. A similar method may also be applied for studying binary, ternary and even quinternary azeotropes.

4. The distillation method gives correct results if we use, for the preparation of a quaternary azeotrope, an excess of those components, which, when mixed with the azeotrope, produce more rapid boiling and condensation temperature increases, than the other ones.

Department of Physical Chemistry, University of Warsaw

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ERRATA AU VOLUME II

Page / Ligne	Au lieu de	Lire
106_{14}	$C_n(x)$	$C(x)$
121_5	bounding	binding
122^1	$\alpha(x_p^{\xi})$	$\alpha(\overset{\xi}{x}_p)$
192_1	values for...	of pyridine found in our previous determination (2) 345.0°, as being correct. As to the critical temperature...
193 Table II	344.60	344.90
194_4	preceding paper	**2** (1954), 77
206^4	$Q_0 \cdot \ldots \cdot Q$	$Q_0 \cdot \ldots \cdot Q_n$
206_{11}	$F_0 =$	$F_0 +$
249_{18}	Re — 41.6%	Re — 41.4%
249_{11}	Re — 41.6%	Re — 41.4%
331^2	n being	we call n
331^4	1.41 ± 2.02	1.41 ± 0.02
331 Table I quatrième colonne	n_E	$\bar{n}_E$
333^5	for high altitude measurements	for measurements at mountain altitudes
334^9	$\gtrsim 10^{12} eV$	$\gtrsim 10^{11} eV$

Note à la communication de A. Wakulicz

Les résultats obtenus par A. Wakulicz sont très voisins de ceux de MM. Pólya et Szegö publiés dans leur livre *Aufgaben und Lehrsätze aus der Analysis* II, Berlin, 1925, pp. 132 et 339, problème 85.